## **Research Group for Molecular Spintronics**

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Spintronics is an emerging technology taking advantage of the dual property of electrons, i.e., the charge and spin degrees of freedom. Over the past decades, spintronics has been developed based on inorganic materials like metals and semiconductors. Recent studies have started to shed light on the spintronics applications of molecular materials including organic molecules and nanocarbons. We call this new field "Molecular Spintronics". In molecular spintronics, the efficient control of the spin direction in molecular materials is essential for device applications by taking advantages of the potentials of these materials for transfer and storage of spin information. Our group focuses on establishing the advantages of molecular materials like graphene by designing the hybrid nanostructures with magnetic materials. For this purpose, the novel spectroscopic and fabrication techniques are employed properly, which allow the microstructure analysis and control at the atomic layer level be possible. We successfully made distinguished to achievements which offer potential spintronic applications of graphene as follows. We are now developing new molecular spintronic devices through graphene-based hybrid structures.

## Spin-dependent electronic properties of graphene/magnetic metal interfaces explored at atomic layer level

Efficient injection of spin-polarized electrons into graphene is the most fundamental issue of graphene-based spin-transport devices. Basically, the spin-injection is performed via the interfaces between graphene and magnetic electrodes, and therefore the understanding and control of the spin-dependent electronic properties of the interfaces are essential for the device design.

In this study [1], we explored the interface properties of graphene and a nickel thin film by utilizing the depth-resolved X-ray magnetic circular dichroism spectroscopy with atomic layer resolution. This innovative approach led to the finding that the contact with graphene induces a dramatic change of spin orientation in the region near the single-layer graphene/nickel interface [Fig. 1].

The depth-resolved analysis of nickel in the single layer graphene/nickel thin film structure demonstrated that the stable spin direction; the easy magnetization direction, changes from in-plane to out-of-plane in the region of a few atomic layers from the single-layer graphene/nickel interface. It was also



Fig. 1 Change of the stable spin direction in the region near the single-layer graphene/nickel interface. The arrows show the spin direction.

shown that the nickel magnetic moment decreases by 20% on the interface in comparison with that in bulk nickel. The analysis of the single-layer graphene showed that the  $\pi$  band of graphene is not only spin-polarized along the out-of-plane direction by the exchange coupling with the interfacial nickel atomic layers but the spin-orbit interaction is remarkably enhanced, which is associated with the  $\pi$ -d interactions at the interface.

In view of the spintronic applications, such interfacial PMA induced by graphene could lead to the small spin-injection efficiency in the grahene-based spin-transport devices with conventional magnetic metal electrodes. On the other hand, the stability of PMA in magnetic ultra-thin films and nanodots is expected to be increased through the interfacial PMA, which is very useful for developing high capacity spin memory devices.

In contrast to the single-layer graphene/nickel structure, we recently demonstrated that the interfacial PMA disappears by replacing single-layer graphene on the interface with single-layer hexagonal boron nitride with a similar honeycomb lattice to graphene. Moreover, our investigations of the single-layer hexagonal boron nitride/nickel structure [2] by utilizing spin-polarized metastable atom deexcitation spectroscopy clarified that single-layer hexagonal boron nitride has a considerably large positive spin polarization in the  $\pi$  band region near the Fermi level [Fig. 2]. This suggests that the single-layer boron nitride contacted with ferromagnetic metals could act as a spin filter which selectively transmits positively spin-polarized electrons and increases the spin injection efficiency in spin-transport devices.

The above spectroscopic investigations of the spin-dependent electronic properties for the interfaces between graphene (and the related materials) and magnetic metals emphasize the importance and usefulness of the graphene/magnetic metal interfaces for designing molecular spintronic devices through the graphene-based hybrid structures.



Fig. 2 Spatial distribution of the spin polarization at the single-layer hexagonal boron nitride/nickel interface in the energy region close to the Fermi level. The orange, blue, and gray spheres show boron, nitrogen, and nickel atoms, respectively. The sign and density of the spin polarization are represented by a gradational change in color (see the color bar on the left side).

## References

[1] Y. Matsumoto *et al.*, J. Mater. Chem. C 1, 5533 (2013).
[2] M. Ohtomo *et al.*, Appl. Phys. Lett. 104, 051604 (2014).